Kondo impurities in nanoscopic systems: new confinement-induced regimes

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We present results for Kondo impurities in nanoscopic systems. Using exact diagonalization in small clusters and Wilson's renormalization group we analyze an isolated system and a nanoscopic system weakly coupled to a macroscopic reservoir. In the latter case, new regimes not observed in macroscopic homogeneous systems are induced by the confinement of conduction electrons. These new confinement-induced regimes are very different depending on whether the Fermi energy coincides with the energy of a resonant state or lies between two quasi-bound states.

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The physics of nanoscopic and mesoscopic systems became of grate interest due to the possibility of making experiments in extremely small samples. Some examples of nano and mesoscopic systems recently studied are small metallic and superconducting islands,[1] quantum dots,[2] nanotubes,[3] and quantum corrals.[4] Depending on the type of system under study, different parameters can be controlled.

The advances in nanotechnologies revived the interest in the Kondo effect,[5] one of the paradigms of strongly correlated systems. On one hand, Scanning Tunneling Microscopy (STM) allowed the direct measurement of local spectroscopic properties of Kondo impurities.[4,6] On the other hand it has been shown that quantum dots and single walled carbon nanotubes weakly coupled to contacts may behave as Kondo impurities generating new alternatives to study the phenomena.[7]

In this work we report results for Kondo impurities in nanoscopic systems. Some experiments show that the effects of confinement are important and recently Affleck and Simon[8] proposed to use a mesoscopic system to measure the Kondo screening length. Although the problem has been addressed theoretically,[9–11] there are still many open questions.[12] What is the temperature dependence of the Kondo screening in nanoscopic systems? What is the effect adding leads or coupling the system to a macroscopic reservoir?

In the case of systems weakly coupled to a macroscopic reservoir, the one electron states acquire a finite lifetime and the spectrum consists of resonant states. This situation is the most relevant to compare with experiments, however due to the intrinsic difficulties to formulate the problem is has not been analyzed in detail. As an example we may mention the case of quantum corrals, were the electrons are not well confined in the corral due to leaking of the wave function and hybridization with the bulk states. Consequently the number of electrons in the corral is not a good quantum number and the local density of states as a function of the energy presents well defined maxima corresponding to the quasi-confined states of the corral.[13] We show that in systems like the quantum corrals new regimes are induced by the confinement of conduction electrons. These new confinement-induced regimes are very different if the Fermi energy is at resonance or between two quasi-bound states. We also discuss the case of quantum dots in Aharonov-Bohm rings.

We have performed numerical diagonalization in small clusters and implemented Wilson's numerical renormalization group (NRG).[14] Our starting point is the Anderson model for magnetic impurities. The model Hamiltonian reads:[15]

$$H_{AM} = \sum_{\sigma} \varepsilon_d d_\sigma^\dagger d_\sigma + U d_\uparrow^\dagger d_\uparrow d_\downarrow^\dagger d_\downarrow + \sum_{\nu,\sigma} \varepsilon_\nu c_{\nu\sigma}^\dagger c_{\nu\sigma} + \sum_{\nu,\sigma} V_\nu c_{\nu\sigma}^\dagger d_\sigma + V_\nu^* d_\sigma^\dagger c_{\nu\sigma} - \mu_i B S_{iz}, \qquad (1)$$

here the operator $d_\sigma^\dagger$ creates an electron with spin σ at the impurity orbital with energy ε_d and Coulomb repulsion U, $c_{\nu\sigma}^\dagger$ creates an electron in an extended state with quantum numbers ν and σ and energy ε_ν. The last term represents the effect of an external magnetic field along the z-direction coupled to the impurity spin $\mathbf{S}_i = d_\sigma^\dagger \tau_{\sigma,\sigma'} d_{\sigma'}$ where τ are the Pauli matrices.

We start by analyzing the case of an isolated system. The properties of the system depend crucially on a few parameters, in particular we show below that in the Kondo limit a finite system presents quite a marked even-odd electron number asymmetry.

We first consider the case of Hamiltonian (1) describing a linear chain of N equivalent sites with the impurity at one end. In this case $\epsilon_\nu = -2t\cos(\nu\pi/(N+1))$, where t is the hopping matrix element in the chain and $V_\nu = V_0 \sin(\nu\pi/(N+1))$ with $\nu = 1, 2, \ldots, N$. We performed exact diagonalization in these finite systems using a Lanczos algorithm.

For an even number of electrons in the system, the ground state is a singlet indicated as $|0\rangle$ and the expectation value of the impurity spin $\langle 0|S_{iz}|0\rangle$ is zero. The impurity spin is screened by the free electrons and the zero temperature susceptibility is finite. We can estimate the characteristic energy scale $k_B T_K$ as the energy involved in the screening of the impurity spin. The magnetization as a function of the external field B, shown in the inset of Fig.1(a), gives an estimation of T_K. We define

$k_B T_K = \mu_i B_c$ where B_c is the crossover field indicated in the inset. As t increases the mean density of conduction states decreases and consequently T_K decreases. As indicated in Fig.1(a) two regimes are obtained: $k_B T_K < \Delta$ and $k_B T_K > \Delta$ where $\Delta \simeq 4t/N$ is the level spacing. For the parameters used the crossover between these two regimes takes place at a hopping $t_{cross} \simeq 0.27$.

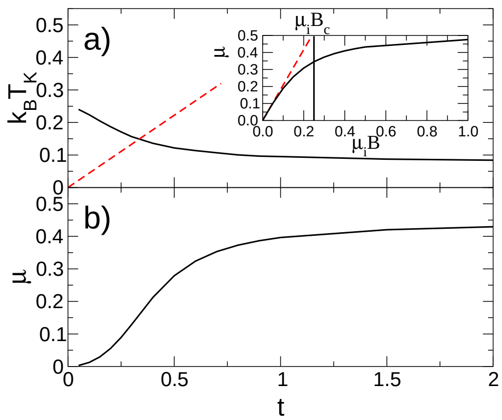

FIG. 1. (a) Kondo temperature, as defined in the text, versus hopping integral t for a chain of nine sites with eight particles ($U = 1.0$, $V_0 = 0.3$, and $\varepsilon_d = \varepsilon_F - 0.5$). The dashed line is the level spacing Δ. Inset: impurity magnetization versus B. (b) Expectation value of the impurity spin versus t, for seven electrons, and the same parameters as in (a).

For an odd number of electrons, the ground state is a Kramers spin-1/2 doublet, $|\Uparrow\rangle$ and $|\Downarrow\rangle$. The expectation value $s \equiv \langle \Uparrow |S_{iz}| \Uparrow\rangle = -\langle \Downarrow |S_{iz}| \Downarrow\rangle$ is different from zero and, in the low temperature limit the impurity susceptibility diverges as $\chi = \mu_i^2 s^2 / k_B T$. In Fig.1(b) the effective magnetic moment $\mu = \mu_i s$ (we take $\mu_i = 1$) is plotted as function of t. For large Δ (large t) the impurity spin is unscreened and $s \approx 1/2$ as expected for a gaped system[16]; for small Δ the impurity spin tends to be completely screened. The crossover between these two regimes takes place at t_{cross}.

Although these results were obtained for a very small system, they give the correct physical picture for zero temperature. While for $\Delta \ll k_B T_K$ the impurity spin is essentially screened regardless of the parity of the electron number, for $\Delta \gtrsim k_B T_K$ an important even–odd asymmetry is obtained. In order to analyze larger systems we have to resort to some approximate scheme. We have adapted the NRG to the present problem. The idea of the NRG is to obtain the quantum many-body states of the system on all energy scales, or length scales, in a sequence of steps. To do that, Wilson defined a basis of concentric wavefunctions in which the Hamiltonian takes the form of a linear chain. A truncated chain with n sites, described by an effective Hamiltonian H_n, gives the correct physics on an energy scale w_n. The NRG transformation $\mathbf{T}$ relates the Hamiltonians describing successive lower energy scales, $H_{n+1} = \mathbf{T}[H_n]$, and leads to a systematic way of calculating the thermodynamic properties at successive lower temperatures.

Going back to our problem and for the sake of simplicity, consider a spherical metallic cluster of radius R_c with the impurity at the center. The cluster is embedded in a bulk material with which it is weakly coupled through a large surface barrier. The Hamiltonian can be put in the form $H = H_{AM} + V(r, R_c)$ where the last term is a potential barrier placed at a distance R_c from the impurity. The Hamiltonian H_{AM} is rewritten in Wilson's basis as schematically shown in Fig.2. The potential barrier is described by including a higher diagonal energy to the $(n_c + 1)$–shell centered around R_c. Alternatively the barrier can be simulated with a smaller hopping matrix element in the region where $V(r, R_c)$ is different from zero. We adopted this last description reducing the hoppings by a factor α.

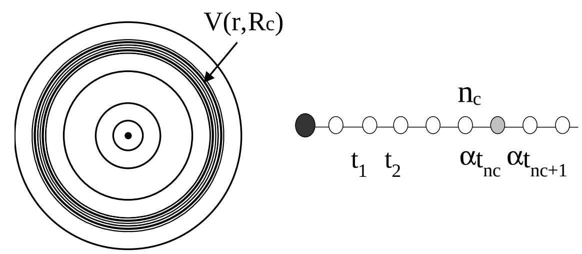

FIG. 2. Schematic representation of Wilson orbitals with the potential barrier and the corresponding linear chain.

If the barrier is impenetrable ($\alpha = 0$), the central cluster is isolated. The process of renormalization ends up after n_c steps and with the obtained low energy states with a *definite number of particles* N_e the partition function Z is evaluated and the thermodynamic properties of the system are obtained. This procedure shows that the high temperature properties, evaluated with a number of shells $n < n_c$, the impurity behaves as in a bulk material. Only when the temperature is lowered and the discrete nature of the one-electron states becomes evident the behavior of the system deviates from that of the macroscopic system. The impurity susceptibility can be evaluated for even and odd number of particles. As we discuss below, to describe even or odd number of particles it is convenient to take even or odd number of shells n_c. The susceptibility is calculated as:[17]

$$\chi = \mu_i^2 \left(\sum_\nu \frac{P_\nu |\langle \nu | S_{iz} | \nu \rangle|^2}{k_B T} + \sum_{\nu \neq \xi} \frac{2 P_\nu |\langle \nu | S_{iz} | \xi \rangle|^2}{E_\xi - E_\nu} \right)$$

where the summation is done over the low energy states $|\nu\rangle$ with energies E_ν and $P_\nu = \exp(-E_\nu / k_B T)/Z$. The matrix elements $\langle \nu | S_{iz} | \xi \rangle$ have to be evaluated in a recursive way at each renormalization step. The susceptibility reflects the thermodynamic properties of the system and we use it as an indicator of the degree of screening of the impurity spin. In Fig.3 the impurity susceptibility is shown for different system sizes. For comparison the same quantity evaluated in the thermodynamic limit

($n_c \to \infty$) is shown. As n_c decreases the characteristic energy separation between the one–electron states Δ increases. In the NRG this energy separation at the Fermi level is $\Delta \simeq D\Lambda^{-n_c/2}$ where D is half the band width and $\Lambda \simeq 2$ is Wilson's discretization parameter.[14]

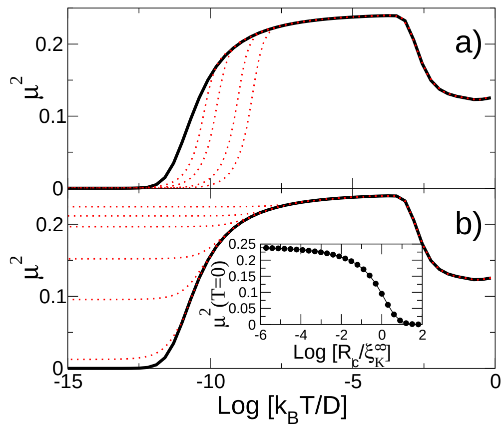

FIG. 3. (a) Impurity magnetic moment squared for a finite system with an even number of particles as obtained with the NRG, the parameters are $D = 1$, $U = 0.02$, $\varepsilon_d = -0.01$, and $V = 0.01$. The thick line corresponds to the thermodynamic limit, thin lines, from right to left, to $n_c = 46$, 50, 56, and 60. (b) Same as in a for (a) system with an odd number of particles, the thin lines from top to bottom correspond to $n_c = 45$, 51, 55, 61, 65, and 71. Inset: Zero temperature impurity magnetic moment squared versus $\log(R_c/\xi_K^\infty)$.

In Fig.3(a) the effective impurity magnetic moment squared[14] $\mu^2 = k_B T\chi$ for an even number of electrons is shown as a function of the temperature. When the temperature is of the order of Δ the behavior deviates from that of the macroscopic sample and μ^2 decreases exponentially due to the gaps induced by confinement. For an odd number of electrons the behavior is illustrated in Fig.3(b). In this case, as $k_B T$ approaches Δ the screening of the impurity spin ends up and a finite magnetic moment survives down to zero temperature, the ground state is a spin-1/2 doublet. In the inset the low temperature magnetic moment versus $\log(R_c/\xi_K^\infty)$ is shown with ξ_K^∞ being the Kondo screening length (see below), the behavior shows the same tendency of that of Fig.1(b) corresponding to exact results of the impurity in a linear chain. Here the crossover between an almost unscreened impurity spin to an almost completely screened spin occurs for $\Delta \simeq k_B T_K^\infty$.

The behavior of the system with a finite barrier is richer and more interesting. Now the central cluster is in contact with a reservoir and the number of electrons in the cluster is not a good quantum number. In the numerical NRG approach the Fermi energy is set to zero. The one particle spectrum in the absence of the impurity has a state at zero energy for even iterations n and two states at approximately $\pm D\Lambda^{-n/2}$ for odd n. Then by taking the barrier enclosing an even or odd number of shells, n_c, we describe a system with the Fermi level at the energy of a resonant state (at resonance) or between two of them (off resonance) respectively. These two situations correspond approximately to an even and odd *average number* of electrons in the cluster respectively.

In Fig.4(a) the impurity magnetic moment μ^2 is shown for the a system at resonance. Only when the temperature is lowered and reaches the value $k_B T \approx \Delta$ the structure in the local density of states becomes important. At low temperatures the universal Kondo behavior is recovered with a Kondo temperature T_K^R higher than T_K^∞, corresponding to a higher density of states. The crossover between the high and low temperature regimes occurs in a small temperature interval leading to a rapid decrease of the magnetic moment μ^2.

For the system off resonance, as $k_B T$ approaches Δ the magnetic moment saturates leading to a plateau in the curve μ^2 versus T as shown in Fig.4(b). Only at lower temperatures the universal Kondo behavior is recovered with a Kondo temperature T_K^{OR} that is lower than T_K^∞. The plateau corresponds to a new regime of partially screened magnetic moment. In this regime, as the temperature is lowered, the thermodynamic properties do not change and are similar to those of a finite system with an even number of particles. The central cluster has a magnetic moment and it is the whole cluster that behaves as a Kondo impurity or a "Kondo grain" when the temperature is lowered. For a given potential barrier height the size of the grain can be changed: systems with small R_c, a large Δ, present a wide plateau with a partially-screened magnetic moment $\mu_{ps}^2 \sim 1/4$. As R_c increases, both the width δT of the plateau where the partially-screened magnetic moment μ_{ps} is stable and the value of μ_{ps}^2 decrease. For a given size, the δT increases with the barrier height as illustrated in Fig.4(c).

In summary, for $k_B T_K^\infty \gg \Delta$ the fine structure (on the scale of T_K^∞) of the density of states does not change the properties of the system and the universal Kondo behavior is recovered. For $k_B T_K^\infty \ll \Delta$ the local density of states, on the scale of T_K^∞, is a smooth function and the universal Kondo behavior is obtained at low temperatures. In this case the system at resonance has a Kondo temperature T_K^R much larger than the corresponding to the system out of resonance T_K^{OR}, however in both cases the high temperature susceptibility is non universal. For $k_B T_K^\infty \sim \Delta$ new confinement induced regimes are observed: for the system at resonance, as the temperature is lowered, there is a rapid decrease in the magnetic moment μ^2 when $k_B T \sim \Delta$; for the system off resonance there is a new partially-screened regime that leads to a plateau in the temperature dependence of μ^2. This plateau can be interpreted as the free-spin regime of the whole Kondo grain that has magnetic moment partially localized at the impurity. At very low temperatures the electrons that are outside the cluster complete the screening. The temperature at which this occurs depends on the coupling between nanoscopic system and the bath. The condition for the existence of these new regimes $k_B T_K^\infty \sim \Delta$ can be put in the form $\xi_K^\infty \sim R_c$ where $\xi_K^\infty = \hbar v_F / k_B T_K^\infty$ is the

Kondo screening length.

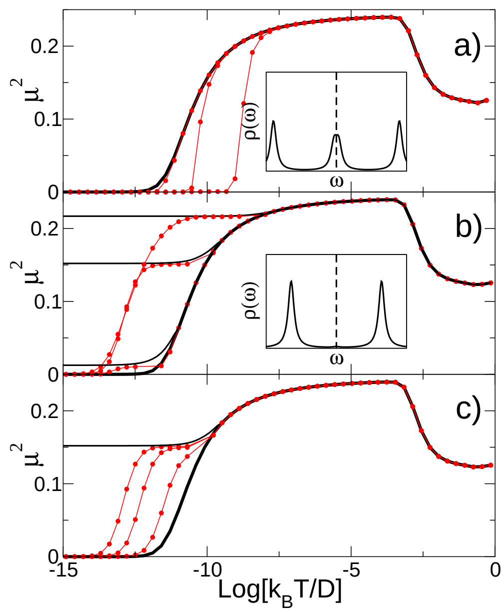

FIG. 4. Impurity magnetic moment squared for a system with a finite potential barrier and the Fermi energy at resonance (a) and off resonance (b) and (c). Parameters are the same as in Fig. (3). In (a) and (b), $\alpha = 0.05$ and different cluster sizes are shown (n_c = 50, 62, 72 in (a) and $n_c = 49$, 61, 71 in (b)). In (c) the potential barrier is changed for a fixed size: $n_c = 61$, $\alpha = 0$, 0.05, 0.1, 0.3, and 1, (from left to right). In (b) and (c) the thin lines that extrapolate to a finite value correspond to the isolated system. In a) and b) the insets show a typical local density of states of the conduction electrons at the impurity site.

For the case of Co impurities in quantum corrals built on the Cu (111) surface, taking $T_K^\infty = 50K$ and using the parameters of the Cu (111) surface band we estimate $\xi_K^\infty \simeq 400\mathring{A}$ which is larger than the typical corral radius. Consequently if the Fermi level is out of resonance we expect the Kondo screening to occur only at very low temperatures. Larger corrals could be built or the effective barrier height decreased by removing some atoms from the corral fence and in this way control the effective Kondo temperature of the impurity.

Our results may have important consequences for the interpretation of experiments with QD in Aharonov-Bohm (AB) rings.[2,7] In the AB configuration, the local density of states inside the ring has the structure of resonant states. A direct consequence of this structure are the oscillations in the transmittance of a ring as a magnetic field threading the circuit is varied. For rings built on GaAs as in some of the experimental setups the energy separation between resonances Δ could be larger than one Kelvin, in fact at 1K oscillations are still observed indicating that at this temperature thermodynamic fluctuations do not wash out the density of states structure. If the Kondo temperature of a quantum dot inserted in one of the ring arms is of the order or smaller than Δ, new effects are to be taken into account. In this case the quantum dot behaves quite differently if the Fermi level lies at resonance or off resonance, corresponding to a maximum or a minimum in the transmittance respectively. The magnetic field can continuously shift the resonances and if the system is in the temperature interval $T_K^{OR} < T < T_K^R$, even at constant temperature the QD continuously evolves from a low temperature regime ($T < T_K^R$) to a high temperature regime ($T_K^{OR} < T$). Moreover, the energy of the resonances depend not only on the external field but also on the phase shift introduced by the QD that is selfconsistently adjusted. On the light of our analysis, in this regime the AB oscillations can not be interpreted directly in terms of a Kondo impurity at a fixed temperature ratio T/T_K.

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